Supplemental material to:

Commuters' Exposure to Particulate Matter Air Pollution is Affected by Mode of Transport, Fuel Type and Route

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In the supplemental material more information is provided on quality assurance of the exposure measurements. Results of laboratory comparison tests between the CPC3007 and CPC3022 are reported. Comparison tests between the three CPC3007 samplers used in the study are described. Comparison tests between the three DataRAMs used in the study are described. Duplicate PM_{10} and soot measurements are discussed.

The supplement also presents additional exposure results. For all modes of transport plots are presented of PNC and $PM_{2.5}$ means of all sampling days. Correlations between pollutants measured in traffic and at the urban background location are presented. In addition to the median concentrations of the air pollutants in all modes of transport in table 1 in the main text, in the supplement also mean of the two-hour mean values and the mean of the two-hour median values are compared to show the effect of peaks on mean and median PNC and $PM_{2.5}$ values. Also more details of the meteorological data are provided.

Quality assurance PNC

We removed PNCs when readings were below 100 pt/cm^3 and when readings changed by more than a factor 10 from one second to the other, because these counts were considered to be unrealistic (Puustinen et al. 2007). In addition, tilts and four seconds after tilts of the instrument were removed. This resulted in removal of 0.3% of the data points, mainly tilts and zero values.
Prior to each measurement in traffic a zero-check was performed using a high efficiency particulate air filter (HEPA).

We performed laboratory tests with artificially generated salt (particles of mean diameter 160 nm) and diesel particles (mean diameter 60 nm) to compare the three CPC3007 devices with a CPC3022, as at particle counts above 100,000 pt/cm³ the CPC3007 is known to underestimate PNCs (Westerdahl et al. 2005; Hameri et al. 2002).
The comparison showed that up to 200,000 pt/cm^3 the CPC3007 measured correctly (Figure 1a). The CPC3007 measures 13% less than the CPC3022, probably because of the difference in lower cut-off particle size (10 vs. 7 nm). For PNCs above 200,000 pt/cm³, the CPC3007 underestimated the CPC3022 PNCs more substantially (Figure1b). We corrected values above 200,000 pt/cm^3 using the following equation: $PNC=83741e^{0,00000458x}$, with x = PNC from CPC3007. This equation was derived by dividing the equation from figure S1b $94669e^{0,00000458x}$ by 1.13 to correct for the 13% difference between the two types of CPCs.
The underestimation in our study was smaller than in the Westerdahl study, possibly related to differences in particle size, distance, or concentration range. Hameri et al reported that CPC3007 counts are reliable up to 100,000 to 400,000 (Hameri et al. 2002).
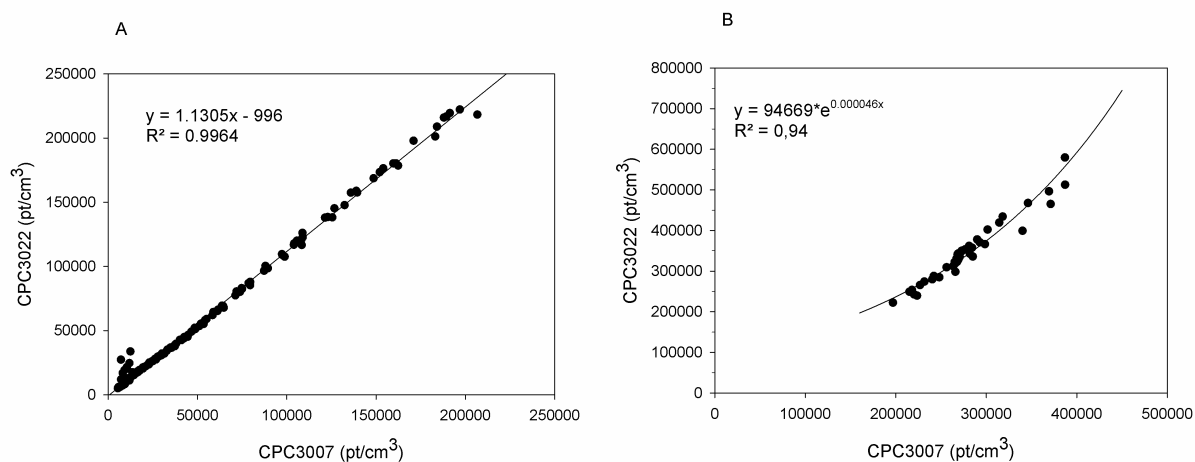
Supplemental Material, Figure 1: Comparison CPC3007 and CPC3022 (laboratory). Data points are minute-averages. (A) Comparison for CPC readings between 5,000 and 200,000 pt/cm³. (B) Comparison for CPC readings between 200,000 and 500,000 pt/cm³.

In the afternoon of each sampling day comparison measurements of the three devices were made. The readings of the three units differed slightly (Figure 2). The differences between the three units changed over time. A moving average of five study days was used as correction factor between the units. The daily correction

factors used to correct two units to the third varied from 0.79 to 1.07. The three units were alternately used in the transport modes and background location, to avoid occurrence of systematic differences between e.g. diesel and electric bus.

A

Y=0.9305x + 955.09
R²=0.9397

B

Y=0.8467x + 106.54
R²=0.9776

C

Y=0.8721 - 355.31
R²=0.9556
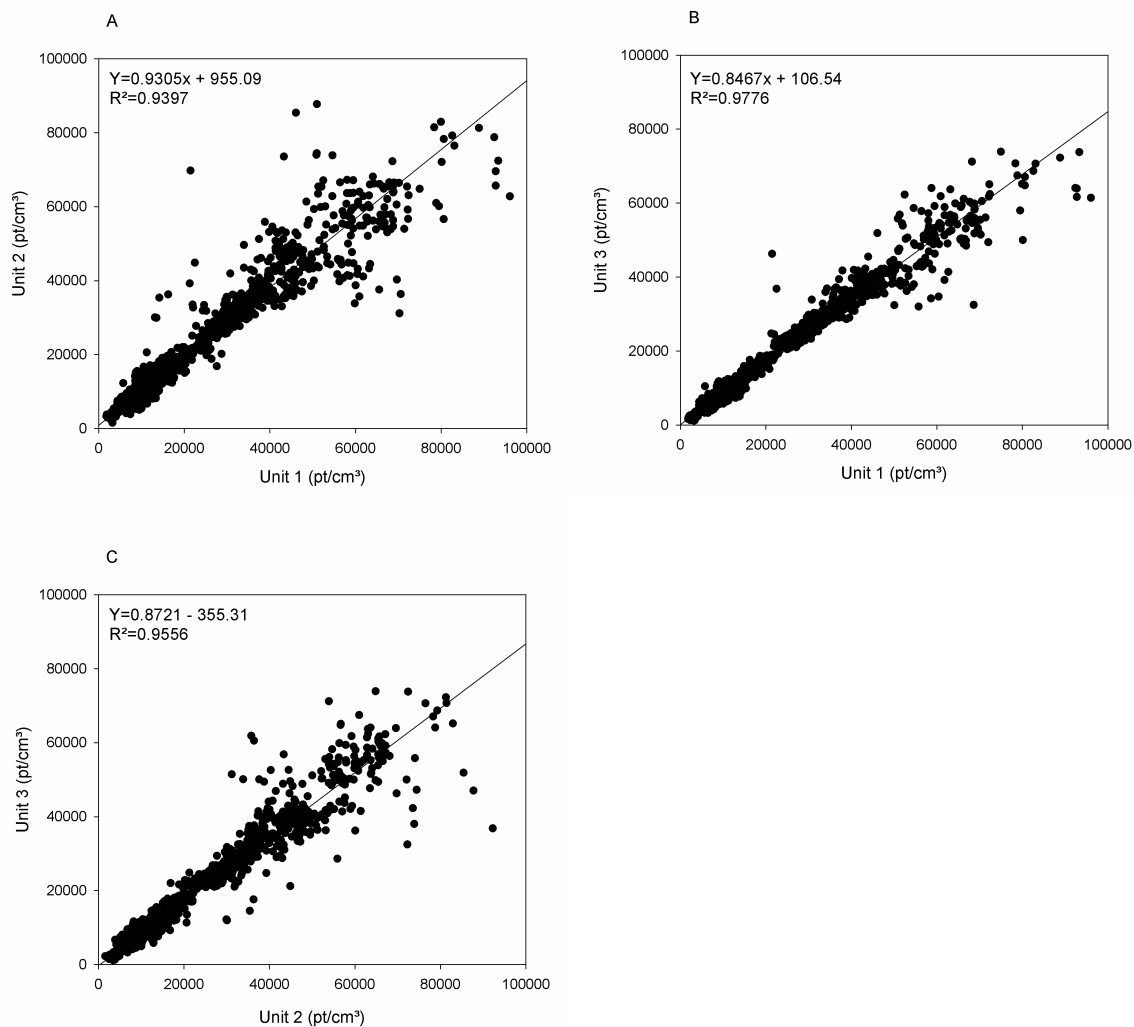
Supplemental Material, Figure 2: Examples of results of comparison measurements of the three CPC3007 used throughout the study. Sampling day December 11th 2007.

Quality assurance PM$_{2.5}$

The DataRAM is based on light scattering technology and readings are known to be influenced by relative humidity (RH). Therefore RH was logged during sampling (Oakton Instruments, IL, USA). Five minute moving averages were used to correct the DataRAM readings with the correction factor:

$$CF = \frac{1}{1 + \frac{0.25 RH^2}{1 - RH}}$$ [1] (Chakrabarti et al. 2004)

Each sampling day comparison measurements were made of the three DataRAMs and five RH loggers used.

The DataRAM factory calibration with ISO Fine test dust was used to translate photometric PM$_{2.5}$ measurements into gravimetric units. This resulted in too high PM$_{2.5}$ levels. Previous studies also reported that PM$_{2.5}$ levels measured photometrically with DataRAMs are 1.4 to 1.9 times higher than measured gravimetrically (Wu et al. 2005;Lanki et al. 2002;Liu, Slaughter, and Larson 2002). Though PM$_{2.5}$ concentrations are likely to be overestimated, relative comparisons between PM$_{2.5}$ exposure in the different modes of transport are considered to be valid. A further limitation is the large impact of the correction factor used to account for the effect of relative humidity on the DataRAM readings, following previous studies (Chakrabarti et al. 2004; Wu et al. 2005). Relative humidity values were frequently in excess of 90%. When leaving PM$_{2.5}$ values out with RH above 90%, differences between buses, cars and bicycles disappeared.

Daily comparison of the three DataRAMs showed small differences between the three units, without a time trend. Study average correction factors of 0.85 and 1.18 were used to correct the readings of two units against the third. Daily comparisons of the relative humidity loggers showed small, varying differences up to 5% (mean: 1.0%) for which no corrections were made.

Quality assurance PM$_{10}$ and soot

We performed measurements in duplicate because of the short sampling time. Before weighing, the filters were equilibrated for 24 hours in stable temperature and RH conditions: temperature varied between 21.6 and 23.2 °C, and RH between 33.3 and 40.3%. An ionising blower was used to minimise effects of static electricity. Pre- and post-weighing of filters were performed in duplicate with a MT5 microbalance (Metler-Toledo Ltd., Greifensee, Switzerland) with one microgram reading resolution.

We used control filters during the weighing sessions. The average weight of 20 field blank filters was subtracted from the sample weights. Based upon precision of measurements, acceptable duplicate criteria for PM$_{10}$ were defined as a weight difference of less than 29 µg or less than 18%. Acceptable duplicate criteria for soot were a difference of less than 2.0 x10^{-5}/m or less than 17% between two filters. If these criteria were not met, both duplicate values were removed. The mean of the duplicate measurements were used in analyses.

We excluded six filters because of pump or power failure. All remaining filter pairs met the duplicate criteria for soot content. Three filter pairs did not meet the duplicate criteria for PM$_{10}$ and were excluded. Two filters with missing duplicates were taken into account because of good duplicate results in the other filter pairs.

The limit of detection (l.o.d.) for PM$_{10}$ and soot, calculated as three times the standard deviation of the field blanks, was 17.4 µg/m³ and 0.95*10^{-5}m^{-1}, respectively. Ten filters (11%) from the background location and one (7%) from a diesel bus were below the l.o.d. of PM$_{10}$;15 filters (16%) from the background location were below the l.o.d. of soot. Values below the l.o.d. were not replaced with a standard value, but retained the measured values.

Mean particle number counts on all sampling days
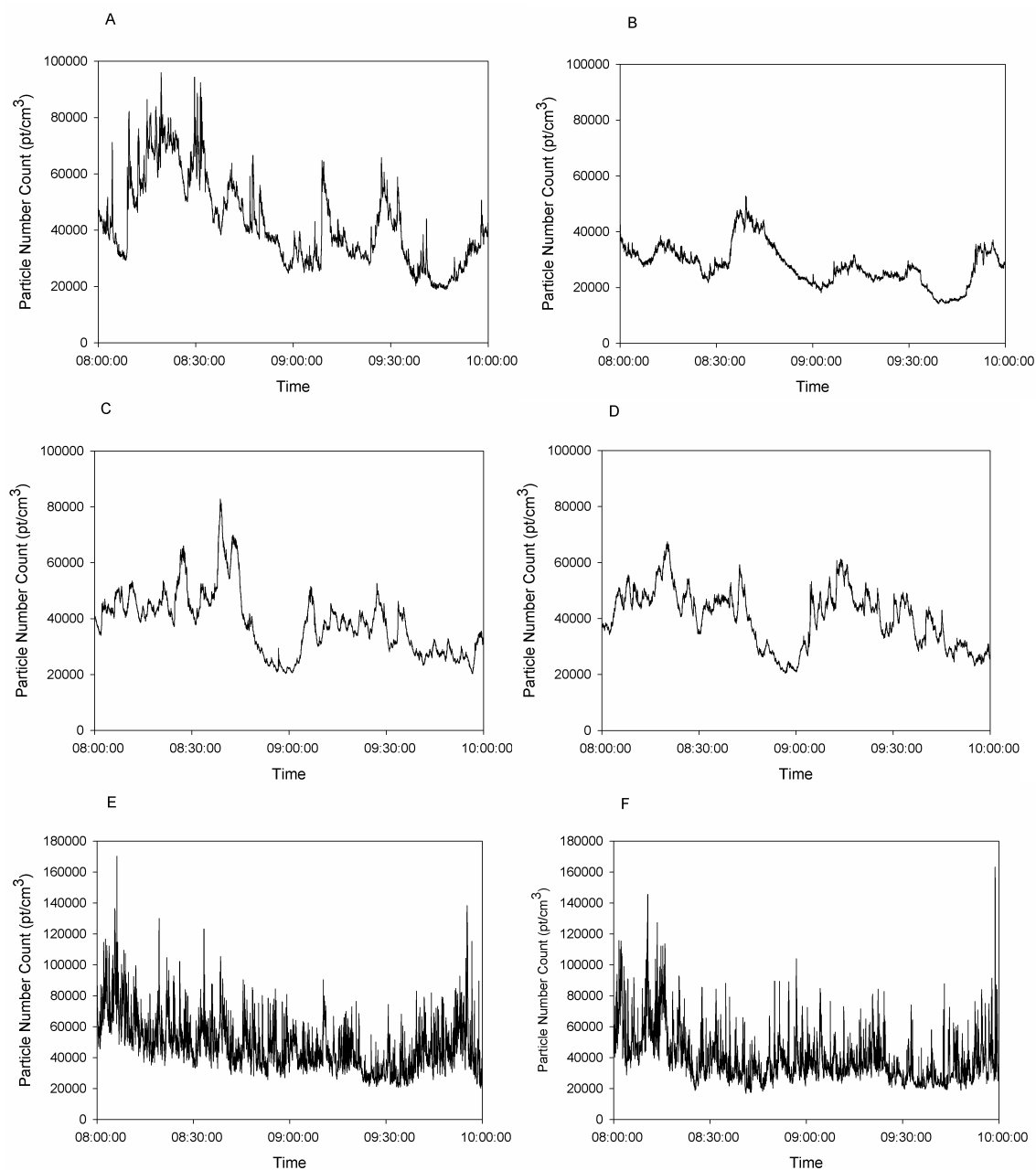
Supplemental Material, Figure 3: Particle number counts; mean of all sampling days. (A) Diesel buses (B) Electric buses (C) Diesel cars (D) Petrol cars (E) High-traffic bicycle routes (F) Low-traffic bicycle routes

6

Mean PM$_{2.5}$ concentrations all sampling days
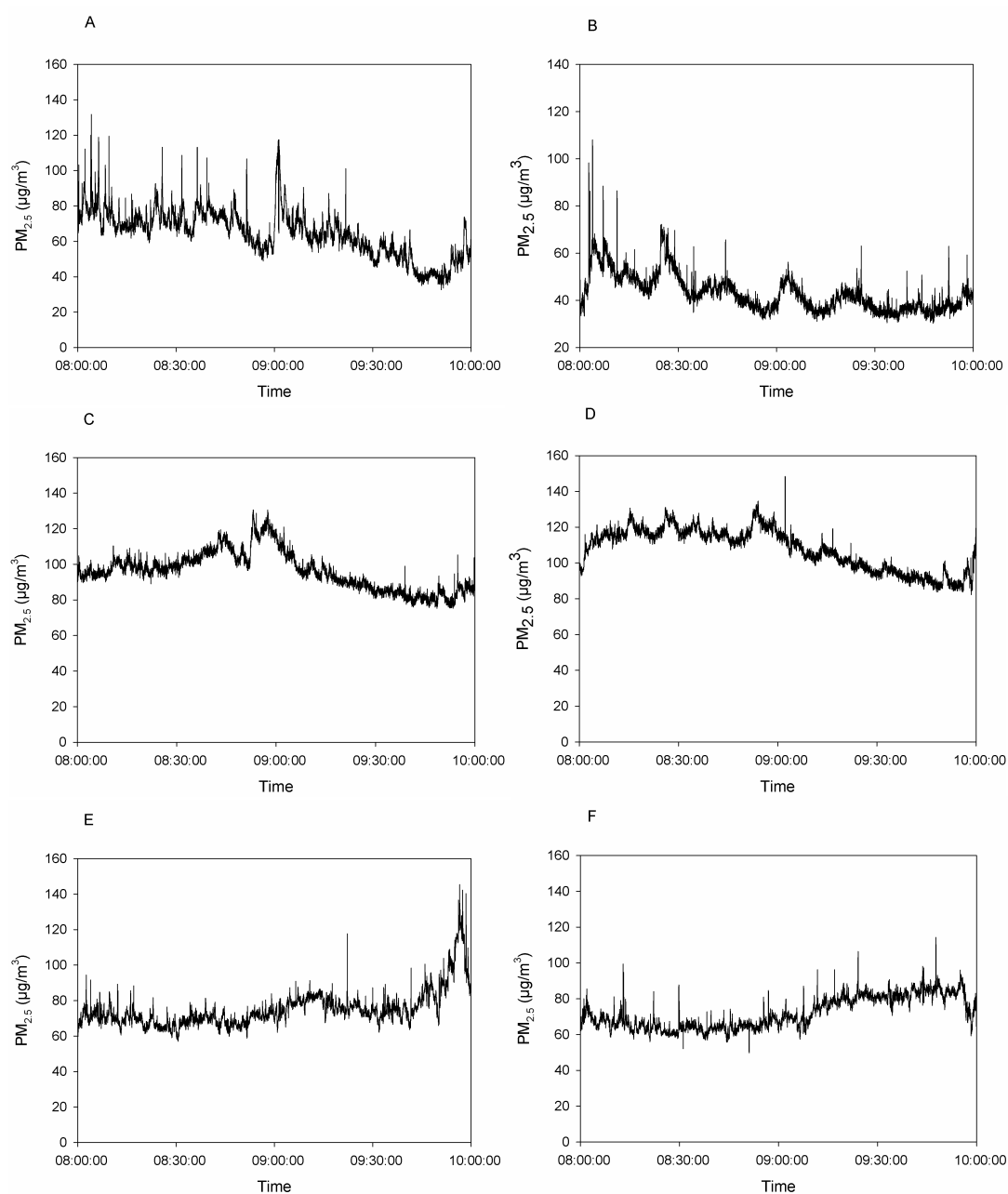
Supplemental Material, Figure 4: PM$_{2.5}$ concentrations; mean of all sampling days. (A) Diesel buses (B) Electric buses (C) Diesel cars (D) Petrol cars (E) High-traffic bicycle routes (F) Low-traffic bicycle routes

Differences in exposure between modes of transport and background

Urban background and commuting concentrations were often significantly correlated, especially for cyclists following the low-traffic route, see Table 1.

Supplemental Material, Table 1: Spearman correlation between concentrations measured during commuting and urban background concentrations

Mode of transport	PNC[a]	PM$_{2.5}$[a]	PM$_{10}$	Soot
Diesel bus	0.01	0.73**	0.69**	0.39
Electric bus	0.49	0.91**	0.38	0.91**
Diesel car	0.60*	0.83**	0.58*	0.68**
Petrol car	0.56*	0.85**	0.27	0.65**
High-traffic bicycle	0.68**	0.93**	0.48	0.84**
Low-traffic bicycle	0.87**	0.82**	0.80**	0.91**

[a])Two-hour mean values, *)Significant, p-level 0.05, **)Significant, p-level 0.01

Table 2 presents means of two-hour mean values and means of two-hour median values. For PNC differences between in-traffic exposure and background exposure are smaller when looking at median values than at mean values, especially for bicycles. This is explained by the many peaks occurring during bicycle rides, that affect mean values more than median values. For PM$_{2.5}$ differences between median and mean values are small, reflecting less peaks in PM$_{2.5}$.

Supplemental Material, Table 2: Difference in mean PNC and PM$_{2.5}$ exposure levels between using two hour mean and two hour median

	n (days)	Mean of mean	Mean of median
PNC (pt/cm^3)			
Diesel bus	13	43235	33557
Electric bus	13	28602	24993
Urban background	13	18908	16961
Diesel car	14	37129	33783
Petrol car	14	40526	30743
Urban background	14	22275	20046
High-traffic bicycle	15	48939	33384
Low-traffic bicycle	15	39576	25087
Urban background	15	23798	22140
PM$_{2.5}$ (µg/m³)			
Diesel bus	10	68.7	64.1
Electric bus	10	40.5	37.3
Urban background	10	33.6	31.7
Diesel car	14	101.3	98.9
Petrol car	14	114.8	111.7
Urban background	14	67.5	66.3
High-traffic bicycle	16	72.3	71.7
Low-traffic bicycle	16	71.7	69.6
Urban background	16	37.8	36.4

Meteorology on sampling days

Average meteorological conditions differed between the three types of sampling days, see Table 3. Rainfall occurred significantly more often on bus sampling days than on car and bicycle sampling days. Wind speed was significantly higher on bus sampling days compared to car sampling days.

Supplemental Material, Table 3: Meteorology on sampling days, mean (SD)

	Bus (n=15)	Car (n=15)	Bicycle (n=16)
Temperature (°C)	7.3 (4.1)	8.4 (6.2)	8.7 (6.7)
Relative humidity (%)	88.7 (8.8)	87.9 (9.7)	84.5 (11)
Air pressure (mbar)	1014 (13)	1016 (9.0)	1015 (10)
Wind speed (m/sec)	4.4 (1.7)	3.3 (1.1)	4.0 (1.9)
Rain (mm)	0.3 (0.7)	0.013 (0.05)	0
Rain 10 hours (mm)*	1.6 (3.0)	0.35 (1.1)	0.17 (0.32)

*Total rainfall between 12 pm and end of commute (approximately 10 am)

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